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- (54) Water-in-volatile silicone emulsion gel cosmetic.
- A water-in-volatile silicone emulsion gel having skin moisturizing qualities is disclosed. The gel is formed by mixing an oil phase with a water phase. The oil phase contains a volatile silicone, a siloxane polyether and an oil. The water phase contains water, a humectant and an oxyethylene functional organosilane. Matching of the refractive index of the oil phase and the water phase results in the formation of a crystal clear gel having a viscosity in the range of 5,000 to 150,000 mm²/s (centistokes).

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This invention is directed to a gel form of personal care cosmetic with moisturizing properties which are beneficial to the alleviation of dry skin conditions.

Inadequate hydration of the stratum corneum results in what is known as dry skin. About ten to twenty percent moisture is the minimum required in order to maintain the retention of normal skin softness and pliability. Accordingly, dry skin sufferers often supplement their own natural skin hydration by the application of consumer products containing humectants and moisturizers. Typically, humectants absorb moisture or aid another substance in the retention of moisture. Moisturizers supply moisture or restore moisture levels in the skin. The goal in either scenario is that of increasing the water content of the skin in order to maintain a soft and pliable skin condition. By including emollients, skin friction is reduced with the result that the skin is enabled to flex without breaking. This is often accompanied by a reduction or retardation in the water loss of the skin.

Of late, the concentration of clear gel forms of personal care products on cosmetic counters is on the rise. Products in clear gel form are very attractive in appearance and are generally regarded as having superior skin absorption characteristics in comparison to conventional emulsion products. Being single phase systems, clear gels have excellent stability. Since consumers perceive such products as being pure and hence safer, there is a growing demand in the marketplace for gel forms of products and especially products with good shelf-life stability which is frequently a problem with clear products.

The water-in-volatile silicone emulsion gel cosmetic of the present invention is a product capable of fulfilling that market need.

The invention introduces a water-in-volatile silicone emulsion gel having skin moisturizing qualities which is formed by mixing together an oil phase and a water phase. The oil phase contains a volatile silicone, a siloxane polyether and an oil. The water phase contains water, a humectant and an oxyethylene functional organosilane. The humectant should be a water soluble material having a refractive index above 1.35. Matching of the refractive index of the oil phase and the water phase results in the formation of a crystal clear gel having a viscosity in the range of 5,000 to 150,000 mm²/s (centistokes).

If the refractive index of the water phase is lower than the refractive index of the oil phase, the indices can be matched by the addition of more of the humectant. If the refractive index of the water phase is higher than the refractive index of the oil phase, the indices can be matched by the addition of more water.

In an alternate embodiment, the humectant is omitted from the water phase and the amount of the oxyethylene functional organosilane is increased. Crystal clear gels have been obtained with this technique in which the oxyethylene functional organosilane also functions as the humectant.

The oil phase of our invention contains a siloxane polyether. One suitable siloxane polyether is a compound having the formula:

$$(R^{a})_{3} - SiO - \begin{bmatrix} R^{a} \\ -SiO \\ R^{a} \end{bmatrix}_{X} \begin{bmatrix} R^{a} \\ -SiO \\ Y \end{bmatrix}_{Y} - Si - (R^{a})_{3}$$

$$(I)_{R^{b}-O-(C_{2}H_{4}O)_{p}-(C_{3}H_{6}O)_{s}-R^{c}}$$

wherein R^a is an alkyl group of one to six carbon atoms; R^b is the radical $-C_mH_{2m}$ -; R^c is a terminating radical which can be hydrogen, an alkyl group of one to six carbon atoms, or an aryl group such as phenyl; m has a value of two to eight; p and s have values such that the oxyalkylene segment $-(C_2H_4O)_p$ - $(C_3H_8O)_s$ - has a number average molecular weight in the range of 400 to 5,000; the segment preferably having fifty to one hundred mole percent of oxyethylene units $-(C_2H_4O)_p$ - and one to fifty mole percent of oxypropylene units $-(C_3H_8O)_s$ -; x has a value of 80 to 120; and y has a value of 2 to 10.

Preferably, R^a and the terminating radical R^c are methyl groups; m is preferably three or four whereby the group R^b is most preferably the radical -(CH_2)₃-; and the values of p and s are such as to provide a number average molecular weight of the oxyalkylene segment -(C_2H_4O)_p-(C_3H_6O)_s- of between 1,000 to 3,000. Most preferably, p and s should each have a value of 18 to 28.

Another suitable siloxane polyether which can be used in the formation of our oil phase is a compound having the formula:

$$(R^{a})_{3} - Sio - \begin{bmatrix} R^{a} \\ I \\ -Sio - \\ I^{a} \\ X \end{bmatrix}_{x} \begin{bmatrix} R^{a} \\ I \\ -Sio - \\ Y \\ R^{b} - O - (C_{2}H_{4}O)_{p} - R^{c}$$
 (II)

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wherein R^a is an alkyl group of one to six carbon atoms; R^b is the radical $-C_mH_{2m}$ -; R^c is a terminating radical which can be hydrogen, an alkyl group of one to six carbon atoms or an aryl group such as phenyl; m has a value of two to eight; p has a value of 8 to 16; x has a value of 6 to 12; and y has a value of 1 to 8.

It should be understood that in either Formula (I) or Formula (II) shown above, the siloxane-oxyalkylene copolymers may take the form of endblocked polyethers, in which the linking group R^b , the oxyalkylene segments and the terminating radical R^c , occupy positions bonded to the ends of the siloxane chain, rather than being bonded to a silicon atom in the siloxane chain. Thus, one or more of the R^a substitutents which are attached to the two terminal silicon atoms at the end of the siloxane chain can be substituted with the segment $-R^b-O-(C_2H_4O)_p-(C_3H_6O)_s-R^c$ or with the segment $-R^b-O-(C_2H_4O)_p-R^c$. In some instances, it may be desirable to provide these segments at locations which are in the siloxane chain as well as at locations which are at one or both of the siloxane chain ends.

Methods of making silicone polyether compounds are known in the art in detail for example, in the volume "Chemistry and Technology of Silicones", Walter Noll, Academic Press Inc., 1968, Pages 373-376.

For the sake of brevity, the siloxane polyether of Formula (I) shall be referred to hereinafter as the "EO/PO Silicone Surfactant", whereas the siloxane polyether of Formula (II) shall be referred to as the "EO Silicone Surfactant".

Often, the "EO/PO Silicone Surfactant" is formulated as a blend containing thirteen percent by weight of the "EO/PO Silicone Surfactant" active ingredient and eighty-seven percent by weight of a volatile cyclic silicone such as octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane or mixtures thereof.

In addition to water and a humectant, the water phase contains an oxyethylene functional organosilane. This water soluble material is a compound having the formula RSiR'₃ in which R is the radical $-O(CH_2CH_2O)_xR''$; R' is an R group or an alkyl radical having one to six carbon atoms; and R'' is a radical selected from hydrogen; an alkyl group of one to six carbon atoms such as methyl, ethyl, propyl and butyl; and an aryl group such as phenyl and benzyl. The integer x can have a value of six to thirty, but is preferably twelve to twenty.

One oxyethylene functional silane particularly preferred has the formula (CH₃)₂Si[O(CH₂CH₂O)₁₆H]₂. Such oxyethylene functional organosilanes may be prepared by methods involving the reaction of a silazane with an organic alcohol in the presence of an inorganic catalyst. One such method is described in detail in U.S. Patent No. 5,157,139.

The oil phase may contain other ingredients in addition to the siloxane polyether such as waxes and emollient oils. Waxes which may be employed include carnauba, beeswax, ceresin, paraffin, candelilla, bayberry, montan, spermaceti, castor wax, ozokerite, microcrystalline waxes and Fisher-Tropsch waxes. Silicone waxes may also be used especially alkylmethylsiloxane wax materials such as polymethylstearylsiloxane. Typically, these silicone waxes are compounds having the formula Me₃SiO(Me₂SiO)_x(MeRSiO)_xSiMe₃ in which Me denotes methyl, R is C_nH_{2n+1} in which n is an integer having a value of at least eighteen, x is 2 to 200 preferably 3 to 70 and y is 3 to 40 preferably 6 to 20. The silicone wax should have a melting point above an ambient or room temperature of 20-25°C. (68-77°F.).

Emollient oils which can be employed in the oil phase include mineral oil, peanut oil, sesame oil, avocado oil, coconut oil, cocoa butter, almond oil, safflower oil, corn oil, cotton seed oil, castor oil, olive oil, jojoba oil, paraffin oil, cod liver oil, palm oil, soybean oil, wheat germ oil, linseed oil and sunflower seed oil; fatty acid esters such as isopropyl myristate, isopropyl palmitate, isopropyl stearate, butyl stearate, cetyl stearate, diisopropyl adipate, isodecyl oleate, diisopropyl sebacate and lauryl lactate; fatty acids such as lauric, myristic, palmitic, stearic, oleic, linoleic and behenic, acid; fatty alcohols such as lauryl, myristyl, cetyl, stearyl, isostearyl, oleyl, ricinoleyl, erucyl and 2-octyl dodecanol, alcohol; lanolin and its derivatives such as lanolin, lanolin oil, lanolin wax, lanolin alcohols, lanolin fatty acids, isopropyl lanolate, ethoxylated lanolin and acetylated lanolin alcohols such as ACETULAN®, a trademark and product of Amerchol Corporation, Edison, New Jersey; hydrocarbons such as petrolatum and squalane; and silicones such as non-volatile siloxane fluids and volatile.

The non-volatile silicones useful in the oil phase are organic polysiloxanes which are film formers having

a viscosity in the range of 5 to several million mm²/s (centistokes), but preferably 100 to 10,000 mm²/s (centistokes). A mixture of polysiloxanes having relatively higher and relatively lower viscosities may be employed. Such polysiloxanes have the repeating unit



 $\begin{array}{c|c}
 & R^1 \\
 & Si \\
 & R^2
\end{array}$

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wherein n is an integer having a value greater than one; R¹ is an alkyl radical containing 1 to 7 carbon atoms or a phenyl group; R² is hydrogen, an alkyl radical containing 1 to 7 carbon atoms or a phenyl group. Illustrative non-volatile polysiloxanes are polydimethylsiloxane, polydiethylsiloxane, polymethylethylsiloxane, polymethylphenylsiloxane, polydiphenylsiloxanes, diphenylsilanediol and copolymers of two or more of the foregoing siloxanes.

The non-volatile silicone film former can be blended with other types of silicone materials such as resins and one particularly preferred silicone film former is a polydimethylsiloxane fluid having a viscosity of one hundred mm²/s (centistokes) measured at 25°C., in admixture with TRIMETHYLSILOXYSILICATE. TRIMETHYL-SILOXYSILICATE is the CFTA adopted name for a silicone resin of the formula [(CH₃)₃SiO_{1/2}]_y in which x and y are positive integers.

Another useful silicone film forming material for use in the oil phase is an ultra-high viscosity silicone gum. These silicone gums typically have the structure HOMe₂SiO(Me₂SiO)_nSiMè₂OH in which Me is methyl and n is an integer having a value of at least one and which can be as much as 10,000.

Because of the high viscosity of these silicone gums and for the purpose of facilitating their handling and processing, such gum materials are often provided in the form of a blend with another volatile or non-volatile low viscosity silicone such as CYCLOMETHICONE or a non-volatile linear silicone fluid having a viscosity of 5 to 350 mm²/s (centistokes). The hydroxy terminated dimethylsilicone polymer has been assigned the adopted name DIMETHICONOL by The Cosmetics, Toiletries and Fragrance Association, Inc., Washington, D.C. (CTFA). Blends of that gum with a volatile low viscosity cyclic silicone have been assigned the adopted name CYCLOMETHICONE (and) DIMETHICONOL by the CTFA. Other blends of such silicone gums with non-volatile low viscosity linear silicones have been assigned the adopted name DIMETHICONE (and) DIMETHICONOL by the CTFA. Typically the DIMETHICONOL content of such blends is 12 to 14 percent by weight and the viscosity of the blended gum may range from 500 to 20,000 mm²/s (centistokes), generally in the range of 4,000 to 5,000 mm²/s (centistokes). The properties of these silicone gums can be modified by replacing one or more of silicone or methyl groups with other substituents such as ethyl and vinyl for example.

The volatile silicone which may be employed in the oil phase is a low viscosity methylsilicone fluid. The volatile low viscosity methylsilicone fluid corresponds to the average unit formula (CH₃)_aSiO_(4-a/2) wherein a is an integer having an average value of from two to three. The methylsilicone fluid contains siloxane units joined by Si-O-Si bonds. Representative units are (CH₃)₃SiO_{1/2}, (CH₃)₂SiO_{2/2}, (CH₃)SiO_{3/2} and SiO_{4/2}. These units are present in molar amounts such that there is provided an average of from about two to three methyl groups per silicon atom in the methylsilicone fluid, whereby the methylsilicone fluid has a viscosity of less than ten mm²/s (centistokes) measured at 25°C.

The volatile low viscosity methylsilicone fluid contains dimethylsiloxane units and optionally trimethylsiloxane units. Representative compounds are cyclopolysiloxane compounds of the general formula $[(CH_3)_2SiO]_x$ and linear siloxane compounds of the general formula $(CH_3)_3SiO[(CH_3)_2SiO]_ySi(CH_3)_3$ in which x is an integer having a value of from three to ten and y is an integer having a value of from zero to four.

The volatile low viscosity methylsilicones have boiling points generally less than 250°C. and possess viscosities generally less than five mm²/s (centistokes) measured at 25°C. Most preferably, the viscosity is 0.65 to 5.0 mm²/s (centistokes).

The cyclopolysiloxane compounds have been assigned the adopted name "CYCLOMETHICONE" by The Cosmetics, Toiletries and Fragrance Association, Inc., Washington, D.C. (CTFA). Both the cyclopolysiloxanes and the linear siloxanes are clear fluids and are essentially odorless, nontoxic, nongreasy and nonstinging. Cosmetically, these methylsilicone fluids are nonirritating to skin and exhibit enhanced spreadability and ease of rub-out when applied. Once applied, the materials evaporate leaving behind no residue.

Volatile methylsilicone fluids which are operable in accordance with the present invention leave substantially no residue after thirty minutes at room temperature when one gram of fluid is placed at the center of a

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No. 1 circular filter paper having a diameter of 185 mm supported at its perimeter in open room atmosphere. By methylsilicone fluid is meant a composition containing two or more silicon atoms, all of which are bonded by way of at least one oxygen atom to at least one other silicon atom and at least one methyl radical, each silicon valence not satisfied by oxygen being satisfied by a methyl radical.

Representative volatile methylsil/cone fluids found to be especially useful in accordance with the present invention are hexamethyldisiloxape which has a boiling point of 99.5°C. and the formula Me₃SiOSiMe₃; hexamethylcyclotrisilexane which has a boiling point of 133°C. and the formula [(Me₂)SiO]₃; octamethylcyclotetrasiloxane which has a boiling point of 171°C. and the formula [(Me₂)SiO]₄; and decamethylcyclopentasiloxane which has a boiling point of 205°C. and the formula [(Me₂)SiO]₅.

These volatile methylsilicone fluids may be used alone or as mixtures in combinations of two or more. Mixtures of the methylsilicone fluids will result in a volatile material having an evaporating behavior different from any one of the individual methylsilicone fluids.

In some instances, it may be desirable to replace one or more of the methyl groups in the volatile methylsilicone fluid with other groups. Thus, there may be substituted groups such as alkyl radicals having two to twelve carbon atoms; aryl radicals having six to ten carbon atoms; amine groups; vinyl; hydroxyl; haloalkyl groups; aralkyl groups; and acrylate groups, for example.

Among the numerous humectants which may be employed as a constituent of the water phase are polyhydroxy alcohols such as sorbitol, glycerin, hexylene glycol, propylene glycol and hexanetriol; sugar and starch derivatives such as alkoxylated glucose, and hydrolyzed mucopolysaccharides; D-panthenol, hyaluronic acid, lactamide monoethanolamine, acetamide monoethanolamine, urea, guanidine, glycolic acid and glycolate salts, lactic acid and lactate salts; and mixtures thereof. Most preferred are glycerin, propylene glycol, urea, hydrolyzed collagen, and sodium pyroglutamate (C₅H₇NO₃Na).

The humectant should be water soluble material having a refractive index above 1.35. Some representative refractive indices are glycerin (1.4744); propylene glycol (1.4325); hydrolyzed collagen (1.4930); and sodium pyroglutamate (1.4295).

In formulating the gel, other adjuvants can be included for the purpose of enhancing the aesthetic appeal of the product such as colorants, preservatives, and fragrances. Only adjuvants which do not effect the stability and clarity of the product should be selected.

Colorants include any of the United States Government Food & Drug Administration (FDA) certified inorganic and organic dyes and lakes such as carmine, iron oxide, mica, titanium dioxide, ultramarines, zinc oxide, bismuth oxychloride; and D & C Blue No. 1, D & C Orange No. 5, D & C Red No. 6 Aluminum Lake, D & C Red No. 7 Calcium Lake, D & C Green No. 8, D & C Red No. 17, FD & C Blue No. 1, FD & C Red No. 3, FD & C Yellow No. 6, External D & C Violet No. 2, which are the CTFA adopted names of The Cosmetic, Toiletry, and Fragrance Association, Washington, D.C.

Preservatives which may be used are methyl paraben, ethyl paraben, propyl paraben, butyl paraben, diazolidinyl urea, imidazolidinyl urea and mixtures thereof. Where an antimicrobial is required, materials such as Triclosan, Quaternium-15, chloroxylenol and cetyl trimethyl ammonium bromide may be employed.

Fragrances which may be used include natural products such as ambergris, benzoin, civet, clove, leaf oil, jasmine, mate', mimosa, musk, myrrh, orris, sandalwood oil and vetivert oil; aroma chemicals such as amyl salicylate, amyl cinnamic aldehyde, benzyl acetate, citronellol, coumarin, geraniol, isobornyl acetate, ambrette and terpinyl acetate; and the various classic family perfume oils such as the floral bouquet family, the oriental family, the chypre family, the woody family, the citrus family, the canoe family, the leather family, the spice family and the herbal family.

In preparing the individual oil phase and the water phase, the oil phase should constitute 10 to 40 percent by weight of the composition, ideally 25 percent by weight. The water phase on the other hand, should constitute 60 to 90 percent by weight of the composition, ideally 75 percent by weight. The oil phase itself is composed of 1 to 20 percent by weight of a volatile silicone; 2 to 20 percent by weight of the siloxane polyether; and 1 to 20 percent by weight of an emollient oil or other film forming material, preferably an organosilicon compound. The water phase should include 5 to 50 percent by weight of water, 1 to 10 percent by weight of the oxyethylene functional organosilane; and 5 to 50 percent by weight of a humectant or other water soluble material having a refractive index above 1.35.

The gel is formed by simply slowly mixing together the two phases under high shear.

The invention may be further illustrated in more detail by a consideration of the following example along with the accompanying table.

Example I

Three crystal clear gel moisturizing compositions "A", "B" and "C" were prepared from an oil phase and a

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water phase containing the various ingredients shown below in the Table. The Table shows the amount of each ingredient in each phase expressed in grams. The ingredients in Oil Phase A were blended and mixed until uniform. The refractive index of Phase A was measured using a refractometer. The ingredients in Water Phase B were blended and mixed until uniform. The refractive index of Phase B was measured using a refractometer. If the refractive index of the water phase was found to be lower than the refractive index of the oil phase, the indices were matched by the addition of more of the humectant. If the refractive index of the water phase was higher than the refractive index of the oil phase, the indices were matched by the addition of more water. Water Phase B was added very slowly to Oil Phase A accompanied with high speed high shear mixing. Crystal clear high viscosity gels resulted having viscosities in the range of 5,000 to 150,000 mm²/s (centistokes). When applied to the skin, the gels possess moisturizing capabilities.

TABLE

Phase	Ingredient	Α	В	С
Α	Volatile Cyclic Silicone	10	10	
Α	EO/PO Silicone Surfactant	10	10	10
Α	Alkylmethyl Silicone Wax	5		
Α	Silicone Fluid/Resin Blend	-	5	5
Α	Silicone Gum	_		10
Phase				
В	Deionized Water	35	35	35
В	Oxyethylene Silane	5	5	5
В	Humectant	35	35	35

In the Table, the humectant was glycerin and each of the silicone and silane materials shown in the Table was a representative compound of one of the categories of organosilicon materials explained previously in detail.

Example II

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Example I was repeated and a fourth crystal clear gel moisturizing composition was prepared having the same formulation as composition "B" in the above Table, except that the humectant was omitted and the level of the oxyethylene functional organosilane was increased to forty. The results were the same as in Example I and a crystal clear gel having skin moisturizing capabilities was obtained.

Claims

- 45 1. A method of making a gel comprising (i) forming an oil phase having a volatile silicone, a siloxane polyether and an emollient; (ii) forming a water phase having water and an oxyethylene functional organosilane; (iii) measuring the refractive index of the oil phase and the water phase; (iv) matching the refractive indices of the oil phase and the water phase; (v) adding the water phase to the oil phase; and (vi) subjecting the phases to high shear mixing.
 - 2. The method of claim 1 further comprising a water soluble humectant with a refractive index above 1.35 in the water phase.
- 3. A method according to claims 1 or 2 in which the volatile silicone is a compound selected from cyclopolysiloxane compounds of the formula [(CH₃)₂SiO]_x and linear siloxane compounds of the formula (CH₃)₃SiO[(CH₃)₂SiO]_ySi(CH₃)₃, in which x is an integer having a value of from three to ten and y has a value of zero to four.

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4. A method according to claims 1 or 2 in which the siloxane polyether is a compound having the formula:

$$(R^{a})_{3} - SiO - \begin{bmatrix} R^{a} \\ -SiO \\ R^{a} \end{bmatrix}_{X} \begin{bmatrix} R^{a} \\ -SiO \end{bmatrix}_{Y} - Si - (R^{a})_{3}$$

$$(I)_{R^{b}-O-(C_{2}H_{4}O)_{p}-(C_{3}H_{6}O)_{s}-R^{c}}$$

wherein R^a is an alkyl group of one to six carbon atoms; R^b is the radical $C_mH_{2m^-}$; R^c is a terminating radical selected from hydrogen, an alkyl group of one to six carbon atoms or an aryl group; m has a value of two to eight; p and s have values such that the oxyalkylene segment $-(C_2H_4O)_p-(C_3H_6O)_s$ - has a number average molecular weight in the range of 400 to 5,000; x has a value of 80 to 120; and y has a value of 2 to 10.

- 5. A method according to claim 4 in which Ra and the terminating radical Rc are methyl groups; m is three; and the values of p and s are such as to provide a number average molecular weight of the oxyalkylene segment -(C₂H₄O)₆-(C₃H₆O)₈- of between 1,000 to 3,000.
 - 6. A method according to claims 1 or 2 in which the emollient is an organosilicon compound selected from (i) an alkylmethylsiloxane wax having the formula Me₃SiO(Me₂SiO)_x(MeRSiO)_ySiMe₃ in which Me denotes methyl, R is C_nH_{2n+1} in which n is an integer having a value of at least eighteen, x is 2 to 200 and y is 3 to 40; (ii) an ultra-high viscosity silicone gum having the formula HOMe₂SiO(Me₂SiO)_nSiMe₂OH in which Me is methyl and n is an integer having a value of 1 to 10,000; and (iii) a silicone film former which is a blend of a polydimethylsiloxane fluid and a silicone resin of the formula [(CH₃)₃SiO_{1/2}k[SiO_{4/2}]_y in which x and y are positive integers.
- 7. A method according to claims 1 or 2 in which the oxyethylene functional organosilane has the formula RSiR'₃ in which R is the radical -O(CH₂CH₂O)_xR"; R' is an R group or an alkyl radical having one to six carbon atoms; and R" is a radical selected from hydrogen; an alkyl group of one to six carbon atoms, an aryl group; and x has a value of six to thirty.
- 8. A method according to claims 1 or 2 in which the oil phase is present in the amount of 10 to 40 percent by weight and the water phase is present in the amount of 60 to 90 percent by weight.
 - 9. A method according to claim 8 in which the oil phase includes 1 to 20 percent by weight of the volatile silicone, 2 to 20 percent by weight of the siloxane polyether and 1 to 20 percent by weight of the emollient oil; and the water phase includes 5 to 50 percent by weight of water, 1 to 50 percent by weight of the oxyethylene functional organosilane and 0 to 50 percent by weight of the humectant.
 - A method according to claim 9 in which the humectant is a compound selected from glycerin, propylene glycol, urea, hydrolyzed collagen and sodium pyroglutamate.
 - 11. A gel obtainable by the method defined in claims 1 or 2.
 - 12. The use of the gel of claim 11 for moisturizing skin comprising applying to the skin an effective amount of the gel and rubbing the gel into the skin.

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EUROPEAN SEARCH REPORT

Application Number EP 94 30 4803

Category	Citation of document with of relevant p	indication, where appropriate,	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Inc.CL6)
X	WO-A-92 05767 (GIL	LETTE)	1-5,8, 10,11	A61K7/48 A61K7/00
	* page 2, line 16 claims 1-23; examp	- page 5, 11me 18; le 1 * 		
A	GB-A-2 217 342 (DO * page 1, line 22 1-15 *	W CORNING) - page 5, line 8; cla	ims 1	
				TECHNICAL FIELDS SEARCHED (Sec.CL.6)
				A61K
	The present merch report has i	area draws un for all claims		
	Place of search	Date of completion of the search	<u> </u>	December
	THE HAGUE	7 December 19	94 WIL	LEKENS, G
X : part Y : part docs A : tuck	CATEGORY OF CITED DOCUME calarly relevant if taken alone calarly relevant if combined with an ment of the same category nelogical background written disclosure	after the file D: document of L: document of	inciple underlying the ot document, but publi ing date lied in the application ted for other reasons the same patent family	***************************************